

## Influence of Hysteresis on Tensile and Fatigue Failure in Rubbers

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### Synopsis

The failure criterion developed by Harwood et al. between energy input to break and hysteresis at break for amorphous rubbers has been related to the fatigue and cut growth properties of the rubber which are based on the tearing energy theory. It is found that the constant  $K$  in the hysteresis failure criterion is a function of the cut growth constant  $G$  and the inherent flaw size  $C_0$ . The effect of adding fine particulate fillers to amorphous rubbers on the hysteresis and fatigue properties is considered and shown to be in agreement with the theory.

### INTRODUCTION

Harwood et al.<sup>1-4</sup> in recent years have found quantitative relationships between the tensile failure of rubbers and the amount of energy dissipated or hysteresis in the polymer, at the point of failure. Extensive studies by NRPA workers<sup>5-7</sup> have shown that fatigue failure of rubber is due to cut growth from small flaws which are initially present in the rubber. The cut growth properties have been explained by use of the concept<sup>8</sup> of "tearing energy." Up to present, these two approaches to the failure of rubbers have been treated separately. This paper, however, attempts to correlate the two approaches and derive a relationship between the parameters from the hysteresis failure criterion with those from the tearing energy theory of fatigue failure.

### TENSILE FAILURE

The energy input to break ( $U_B$ ) of a rubber when subjected to a tensile test is related to the energy dissipated or hysteresis at break ( $H_B$ ) represented by the area of a stress-strain loop just before the point of failure (omitting a small temperature correction) by

$$U_B = K(H_B^{2/3}) \quad (1)$$

This has been found<sup>2,3</sup> to hold for a number of amorphous rubbers over a temperature range of  $-40^\circ\text{C}$  to  $140^\circ\text{C}$ . If, however, eq. (1) is written in

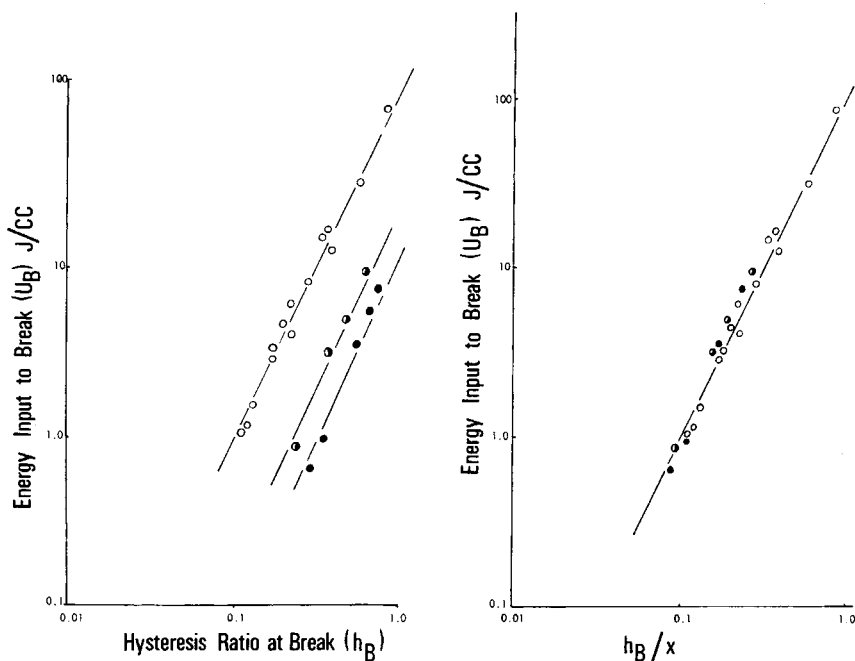


Fig. 1. (a) Variation of energy input to break with hysteresis ratio at break for NBR gum and NBR containing 60 and 80 phr HAF carbon black. Lines through points follow a square law as predicted by eq. (2). (b) Variation of energy input to break with hysteresis ratio at break corrected by  $X$  factor, as predicted by eq. (15), for NBR gum and NBR containing 60 and 80 phr HAF carbon black: (O) NBR gum; (◐) NBR + 60 phr HAF; (●) NBR + 80/phr HAF.

terms of hysteresis ratio at break ( $h_B$ ) (i.e., the ratio of the hysteresis at break,  $H_B$ , to the energy input to break,  $U_B$ ), the equation becomes

$$U_B = K^3 h_B^2. \quad (2)$$

This is shown in Figure 1a for acrylonitrile-butadiene rubber (NBR). The value of the constant  $K^3$  has been found to vary slightly between different polymers. A recent paper<sup>3</sup> has suggested that  $K^3$  is the value of energy input to break at the glass transition temperature of the polymer and is thought to reflect the cohesive energy density of the rubber.

### FATIGUE FAILURE

The fatigue life of rubber when subjected to repeated deformations is dependent on a parameter termed tearing energy ( $T$ ). Tearing energy<sup>8</sup> is defined as the elastic strained energy dissipated per unit area of crack growth and can be expressed by

$$T = \left( \frac{\partial U}{\partial A} \right)_e \quad (3)$$

where  $U$  is the total strain energy in the testpiece and  $A$  is the surface area of the crack measured in the unstrained state. The suffix  $e$  denotes that the differentiation is carried out at constant deformation.

It has been shown that a minimum value of tearing energy ( $T_0$ ) exists below which there is no mechanical cut growth, and hence this defines a fatigue limit for repeated stressing below which the life can be indefinite in the absence of any chemical effects. It has also been shown<sup>3,11</sup> that  $T_0$  corresponds to the lower limit of energy input to break values on the hysteresis-at-break graph shown in Figure 1a where the rubber is considered to be in a totally elastic state.

Above  $T_0$ , it has been found<sup>5-7</sup> that the amount of cut growth per cycle ( $dc/dn$ ) for a tensile strip test piece containing an edge crack in a repeated extension test can be expressed by an equation of the following form:

$$\frac{dc}{dn} = \frac{T^m}{G} \quad (4)$$

where  $G$  is a constant, and this is illustrated for a number of rubbers in Figure 2. The actual value of the power  $m$  is unfortunately dependent on the type of polymer<sup>7</sup>; for natural rubber,  $m = 2$ ; for a range of synthetic rubbers (e.g., butyl, polychloroprene, polybutadiene, PVC),  $m = 3$ ; and for styrene-butadiene rubber,  $m = 4$ . This type of equation has also only been shown to hold for measurements carried out at room temperature (approx. 21°C).

For a test piece in the form of a strip with a cut of length  $C$  along its edge, the tearing energy  $T$  is given by

$$T = 2kUC \quad (5)$$

where  $U$  is the strain energy density in the bulk of the test piece remote from the cut and  $k$  is a slowly varying function of strain<sup>12</sup> which has a numerical value of 2 in most cases.

If it is assumed that  $m = 3$ , which is normally the case for amorphous polymers, then  $T$  can be eliminated by use of eqs. (4) and (5) to yield

$$G \frac{dc}{dn} = T^3 = (2kU)^3 C^3. \quad (6)$$

This differential equation can be solved to give the number of cycles  $n$  to increase the crack from  $C_0$  to  $C_1$ :

$$n = \frac{G}{2(2kU)^3} \left[ \frac{1}{C_0^2} - \frac{1}{C_1^2} \right]. \quad (7)$$

When failure occurs, the final crack length will be much greater than the initial flaw size  $C_0$ , and hence the number of cycles to failure  $N(e)$  is given by

$$N(e) = \frac{G}{2(2kU)^3} \frac{1}{C_0^2}. \quad (8)$$

Equation (8) expresses the fatigue life  $N(e)$  of a rubber when subjected to continuous cycling at constant temperature but at varying strains ( $e$ ) in terms of the cut growth constant  $G$ , initial flaw size  $C_0$ , and maximum strain energy  $U$ , which can be derived from the maximum strain. Good agreement has been found<sup>6</sup> between the predicted fatigue life from eq. (8) and ex-

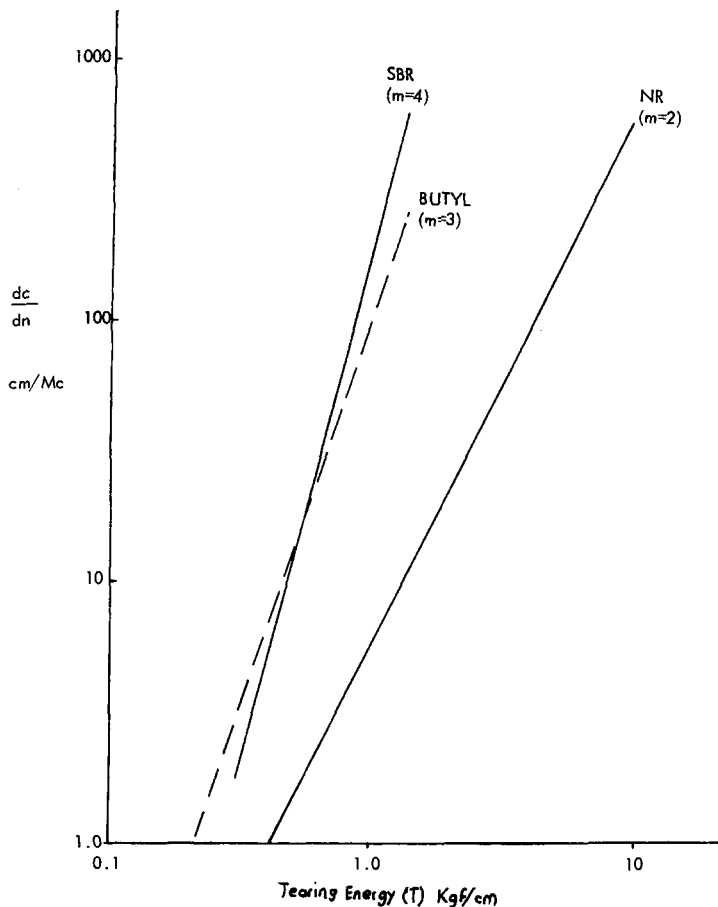


Fig. 2. Variation of cut growth per cycle with tearing energy for a number of rubbers, from data of Lake and Lindley.<sup>7,9</sup>

perimental data provided that the correct value of  $m$  for each particular polymer considered is substituted in the equations.

Recent work<sup>13,14</sup> has also shown that if fatigue tests are carried out on synthetic rubbers at a fixed strain but over a variable temperature range, then the fatigue life is dependent on the amount of hysteresis exhibited by the rubber at that strain. For SBR at 175% extension and over a tempera-

ture range of  $-40^{\circ}\text{C}$  to  $+80^{\circ}\text{C}$  and over a frequency range 0.008 Hz to 8 Hz, the fatigue life  $N(t)$  can be expressed as

$$N(t) = \frac{h^p}{A} \quad (9)$$

where  $h$  is the hysteresis ratio raised to the power  $p$ , which was found approximately to be 6, and  $A$  is a constant.

### RELATION BETWEEN FATIGUE AND STRENGTH/ HYSTERESIS THEORY

If it is assumed that the total fatigue life  $N$  over a variable temperature range  $T$  and range of strain  $e$  can be expressed as

$$N = N(e) \cdot N(t), \quad (10)$$

then a relationship for total fatigue life ( $N$ ) can be obtained by combining eqs. (8), (9), and (10) to yield

$$N = \frac{G}{2(2k)^3} \frac{1}{AC_0^2} \frac{h^6}{U^3} \quad (11)$$

If  $N = 1$ , which is the case with a normal tensile test, then from eq. (11)

$$U = \left( \frac{G}{2(2k)^3 AC_0^2} \right)^{1/3} h^2. \quad (12)$$

The form of eq. (12) is the same as the hysteresis failure criterion found from tensile tests shown in eq. (2). The constant  $K^3$  in eq. (2) is therefore identified with constants derived from the fatigue failure equations:

$$K^3 = \left( \frac{G}{2(2k)^3 AC_0^2} \right)^{1/3}. \quad (13)$$

If it is assumed that the parameter  $A$  remains constant for a number of amorphous polymers, then for rubbers obeying eq. (6),  $K^3$  is proportional to  $(G/C_0^2)^{1/3}$

Values of  $K^3$  from the data of Harwood and Payne<sup>3</sup> and values of  $C_0$  and  $G$  from the data of Lake and Lindley<sup>7,9</sup> are shown in Table I.

The variation of  $K^3$  with  $G/C_0^2$  is shown in Figure 3 for the rubbers listed in Table I with the exception of NR, which crystallizes strongly on extension and does not obey either eq. (2) or (6). A line of slope  $1/3$  predicted by eq. (13) is shown in Figure 3 to be a reasonable approximation to the results. Apart from experimental error, the difference between the experimental points and the  $1/3$  power law can be explained by (a) the slightly different mixed compounds used by Harwood and Payne and Lake and Lindley, (b) the likely variation in the constant  $A$  between different polymers; (c) for some rubbers,  $m = 3$  is only an approximation, and for SBR in particular, a value of  $m = 4$  in eq. (4) should be used.

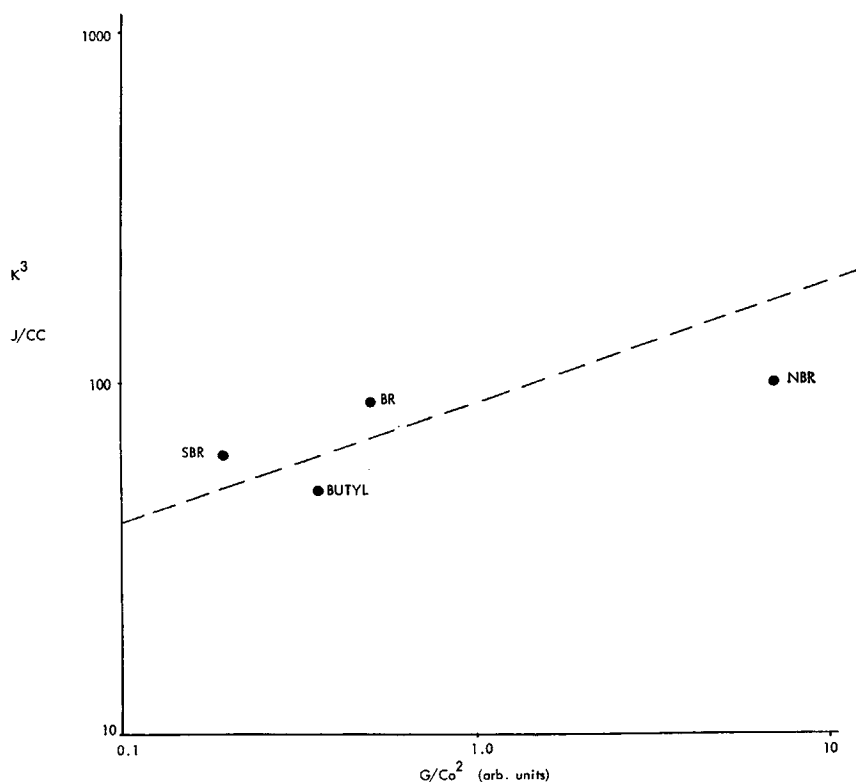


Fig. 3. Relationship between  $K^3$  from eq. (2) and  $G/C_0^2$  from eq. (12). Dotted line of slope  $1/3$  predicted by eq. (13) is also shown. Actual values are listed in Table I.

The hysteresis failure criterion, eqs. (1) and (2), however, is obeyed for SBR as well as for all other amorphous rubbers and also to a limiting extent for a strain-crystallizing rubber such as natural rubber; and this possibly indicates the usefulness of the equation compared with fatigue equations which vary considerably between the different polymers.

TABLE I  
Comparison of Tensile and Fatigue Failure Parameters

Rubber	$K^3$ , <sup>a</sup> J cm <sup>-3</sup>	$C_0$ , <sup>b</sup> cm × 10 <sup>3</sup>	$G$ , <sup>c</sup> arb. units	$G/C_0^2$ , arb. units
Butyl	49	5	0.091	0.036
SBR	61	5.5	0.056	0.019
BR	88	2.5 <sup>d</sup>	0.031	0.050
NBR	97	4	1.100	0.690
NR	124	2.5	2.200	3.500

<sup>a</sup> From Harwood and Payne.<sup>3</sup>

<sup>b</sup> From Lake and Lindley.<sup>9</sup>

<sup>c</sup> From Lake and Lindley.<sup>7</sup>

<sup>d</sup> Assumed same as NR as shows evidence of strain crystallization on extension.

## EFFECT OF FILLERS

The addition of fine particulate fillers such as carbon black to amorphous rubbers increases their modulus and strength. The hysteresis failure criterion, eq. (2), has been found to hold for filled rubbers containing carbon black.<sup>1,3,4</sup> The failure lines of the filled rubbers are displaced away from the unfilled rubbers as shown in Figure 1a for NBR containing 60 and 80 phr HAF carbon black. The displacement of the filled rubbers away from the gum rubber increases with increasing concentration of carbon black. This additional loss of energy arises because the filler particles are inextensible and the overall strain of the sample is less than the strain in the rubber phase. Mullins and Tobin<sup>15</sup> have suggested that in filled rubbers the modulus of the rubber phase is increased by a factor  $X$  which takes account of both the disturbance of the strain distribution and the absence of deformation in that fraction of the composite composed of filler. They considered the relationship.

$$X = \frac{\sigma}{eE_0} = \frac{E}{E_0} = 1 + 2.5c + 14.1c^2 \quad (14)$$

where  $e$  is the strain produced by the stress  $\sigma$ ,  $E_0$  is the modulus of the rubber without filler, and  $c$  is the volume concentration of filler. This expression is identical to the hydrodynamic equation developed by Guth and Gold<sup>16</sup> relating the viscosity of a liquid containing hard spherical particles to that of the liquid alone.

It was found that by dividing the hysteresis ratio axis by  $X$ , the results from the unfilled and filled vulcanizates coincided as shown in Figure 1b, and hence eq. (2) can be modified to take account of both filled and unfilled rubbers as shown in eq. (15) when the small temperature correction factor is omitted:

$$U_B = K^3 \left( \frac{h_B}{X} \right)^2. \quad (15)$$

For filled rubbers it can be considered, therefore, that  $K^3$  is reduced by the factor  $X^2$ .

It has been suggested<sup>14</sup> that eq. (9) holds for both filled and unfilled rubbers. Limited work has been done<sup>7,9</sup> on the fatigue properties of filled rubbers; but it has been shown, that the "effective" initial flaw size  $C_0$  is larger for filled rubbers than gum rubbers when calculated back from fatigue data. In one particular case quoted,<sup>9</sup> the initial flaw size doubles from  $2.5 \times 10^{-3}$  cm to  $5.0 \times 10^{-3}$  cm by the addition of 50 phr HAF carbon black to natural rubber. The experimental results show therefore that when carbon black is added to rubber, the value of  $K^3$  is reduced by the hydrodynamic factor and the effective initial flaw size is increased. If it is assumed, therefore, that  $A$  remains constant, these two experimental observations are in qualitative agreement with that predicted in eq. (13).

## CONCLUSIONS

It has been shown that the failure criterion between hysteresis at break and energy input at failure found by Harwood et al. can be derived from equations used to predict the fatigue behavior of rubbers. The constant  $K^3$  in the hysteresis failure criterion has been identified with constants from the fatigue behavior, the most important of which is the initial flaw size. In contrast to the fatigue equations which show different power law relationships for different rubbers, all conventionally compounded amorphous rubbers are found to obey the same hysteresis failure criterion, which indicates the importance of this relationship.

The variations in both tensile and fatigue behavior when carbon black is added to rubbers is shown to be in accord with the general theory. It is likely that the observed increase in "effective" initial flaw size with filler loaded vulcanizates is due to a hydrodynamic factor such as the one derived by Guth and Gold shown in eq. (14), which has already been successfully applied to a number of properties of filled rubber vulcanizates.

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